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Optimisation of application of acid protease and transglutaminase on wool to achieve machine washable care claim

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Acid protease and transglutaminase enzymes have been used to impart anti-felting properties to wool with minimal loss in tensile strength. Statistical optimisation of the application conditions using Box Behnken design of experiment has been employed to achieve less than 3% total warp shrinkage as specified in Woolmark Specification AW-1 for machine washable care claim. During optimisation of protease treatment, the total warp shrinkage is reduced from 7.66% to 2.77% but is accompanied with 9.95% loss in warp tensile strength. Subsequently, optimisation of transglutaminase enzyme has been carried out on protease treated wool. This results in marginal reduction of total warp shrinkage to 2.64% with 7.92% recovery in warp tensile strength, giving a net loss of 2.82% in warp tensile strength.

Keywords: Acid protease, Anti-felting property, Machine washable, Shrink-resist, Transglutaminase, Wool

1 Introduction

Wool's natural tendency to shrink during washing significantly impacts end-product performance. Its complex morphological structure includes the cuticle (10% of wool weight) and cortex (90%)[1]. The cuticle, comprising the epicuticle, A-layer, B-layer, and endocuticle, surrounds the cortex. The epicuticle membrane contains around 25% fatty acid, primarily 18-methyleicosanoic acid, giving wool a hydrophobic surface[2]. Cationic detergent treatments can remove up to 65% of this surface bound fatty acid[3]. The cuticle surrounds compacted cortical cells aligned with the fibre axis, separated by the cell membrane complex, ensuring strong intercellular bonding via proteins. The cuticle has a higher degree of cysteine crosslinks than the cortex[4-6]. The peculiar arrangement of the cuticle scales leads to high degree of felting shrinkage exhibited during laundering. Decades of research have focused on developing processes to counteract wool shrinkage, leading to successful commercial techniques utilizing various chemicals for creating machine-washable products. Chlorine-based methods, such as the Chlorine-Hercosett process, dominated, with modifications explored, like combining Hercosett polymer and anionic surfactant on thioglycolate-treated wool[7-8]. Early processes caused damage, resulting in tendering, weight loss, and yellowing. Extensive research improved treatment control, introducing alternative chemicals like dicholoroisocyanuric acid, potassium peroxymonosulphate, potassium permanganate, often combined with polymers (Basolan SW, Basolan MW, Hercosett polymer, Polymer RSM, GE) to reduce felting shrinkage and enhance fibre softness[9-10]. Another approach involves inter-fibre crosslinks, utilizing products like Synthappret LKF, Synthappret BAP, silicones, and self-crosslinking polyacrylates, though some processes caused damage or made fibres stiff and harsh[11]. The effect of proteolytic and lipolytic enzymes on untreated and chemically shrink resisted wool was studied. It was observed that the chemically shrink-resisted wool was more susceptible to damage by the enzymes as compared to untreated wool[12].

Due to environmental concerns associated with chlorine-based compounds, a shift towards eco-friendly processes became imperative. Enzymes, particularly protease enzymes, were introduced to address wool felting shrinkage. Papain, in combination with lipase and sodium monoperoxyphthalate, reduced felting shrinkage from 32.6% to 1.8%, with a 12-13% strength loss and 4.86% weight loss[13]. Acid and alkaline proteases were compared, with acid peroxide bleaching followed by acid protease treatment showing lower weight (1.84%) and strength loss (8.47%) than alkaline peroxide bleaching with alkaline protease treatment

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(6.45% weight loss and 19.04% strength loss). Acid peroxide bleaching with subsequent acid protease treatment imparted higher felting shrinkage (7.77%) as compared to alkaline peroxide bleaching followed by alkaline protease treatment (3.64%) but with better softness rating of 4.4 as compared to 3.4 in case latter. Alkaline peroxide bleaching with subsequent acid protease treatment yielded intermediate results (weight loss of 2.34%, strength loss of 9.34% and felting shrinkage of 5.23%) with higher softness rating (4.6)[14]. Bromelain treatment at pH ~6 resulted in <6% area shrinkage, 7.56% weight loss, 9.53% strength loss, and a 3.7 softness rating[15]. Other studies also indicated proteolytic activity of acid proteases on wool[16,17]. The effect of hydrogen peroxide and dicholorodicyanuric (DCCA) acid pretreatmentswas compared. It was observed that DCCA pretreatment followed by protease treatment gave remarkable improvement in felting shrinkage, whereas hydrogen peroxide pretreatment followed protease treatment did not show any remarkable improvement in antifelting properties[18]. Many other types of enzymes were explored including lipases, isomerases, oxidases and peroxidases. Various enzymes, including lipases, isomerases, oxidases, and peroxidases, were explored, with protease treatment notably enhancing fibre softness and dye absorption[19,20]. Thermo- and alkali-stable proteases from extremozymes organisms improved whiteness, dyestuff uptake, and reduced felting tendency, albeit with a decrease in colour fastness[21]. The addition of protease to an alkaline peroxide bleaching bath, followed by chitosan biopolymer application, enhanced whiteness, wettability, and machine washability of wool[22]. Among 16 proteases, Esperase was found most effective in conferring shrink-resistance, especially when pre-treated with alkaline sodium bisulphite[23].

Protease treatment, known for causing a significant degree of strength loss, particularly with acid proteases demonstrating lower strength loss, was attributed to damage to the cortex by these enzymes. To mitigate this, efforts were made to limit the action of protease enzymes to the scales by immobilizing them. Using native and immobilized Esperase Protease to treat wool resulted in 25% and 8% strength loss, respectively, with an area shrinkage <5% in both cases[24]. Similarly, other studies using immobilised enzyme showed better results as compared to native proteases[25,26]. To prevent the diffusion of protease enzymes into the interior of wool fibres and reduce strength loss, a high concentration of salt was employed during treatment[27]. Increasing the molecular weight of proteases by covalently attaching PEG resulted in modified proteases retaining 80% activity and causing 90% less weight loss, with a slight reduction in felting tendency[28]. Covalently binding subtilisin to PEG revealed that bound subtilisin selectively hydrolysed the cuticle layer of wool, leading to higher tensile strength and lower felting of the fibre compared to free subtilisin[29].

The second strategy used transglutaminase to remediate damage to wool and recover the lost strength. Tissue transglutaminase and microbial transglutaminase were compared, with tissue transglutaminase showing slightly better strength recovery, while microbial transglutaminase performed slightly better in reducing felting shrinkage[30]. However, transglutaminase alone could not significantly improve felting shrinkage and tensile strength of untreated wool due to the hydrophobic fatty layer. Various pretreatments were explored to counteract this layer, and treatment with alkaline protease (Savinase 6L) reduced area shrinkage from 9.5% to 7% with a 19.4% loss in tensile strength. Subsequent transglutaminase treatment resulted in an 80% recovery in tensile strength with a reduction in area shrinkage to 5.4%[31]. The combined effect of hydrogen peroxide, Savinase 16L, microbial transglutaminase, and their combination was studied. Hydrogen peroxide treatment followed by protease treatment reduced felting shrinkage to 6%, with high alkali solubility of 23% and a 25% loss in tensile strength. Subsequent transglutaminase treatment resulted in a 12-16% improvement in tensile strength with a 2-4% decrease in alkali solubility[32]. Air plasma, Savinase 16L, and microbial transglutaminase enzyme treatment were explored to modify knitted wool fabric, with air-plasma treatment resulting in an insignificant strength loss of 1.5%. Subsequent protease treatment led to a 9% loss in strength, and transglutaminase treatment increased tensile strength, resulting in a 2% net loss in tensile strength. Area shrinkage reduced from 17.2% to 10.47% with air-plasma treatment, to 3.4% with protease treatment, and to 2.3% with subsequent transglutaminase treatment[33]. Transglutaminase was also utilized to modify the wool surface by incorporating different proteins, such as casein and keratin hydrolysates, resulting in reduced area shrinkage and increased tensile strength. Additionally, transglutaminase was

employed for grafting silk proteins onto wool, leading to increased bursting strength, reduced felting shrinkage, and improved fabric softness. Finally, transglutaminase treatment increased wool's resistance to damage by both biological and non-biological detergents, preserving its strength better[34-38].

In this study, an acid protease and microbial transglutaminase from *Streptoverticilium mobaraense* have been used to treat wool to reduce felting shrinkage and remediate the damage caused by the protease treatment. The conditions of the application for this simultaneous application are optimised using Box-Behnken design of experiment.

2 Materials and Methods

2.1 Materials

Pure wool fabric, possessing 2/64s in both warp and weft, EPI 81, PPI 64 and GSM 185, was used. The wool fabric was scoured with Sarafree-S (1.0%) at 40-45°C, *p*H 6.5 for 30min. Subsequentlyit was dried at 150°C and open decatised.

Acid protease (AP) and microbial transglutaminase (TG) enzyme were sourced from Rajvi Enterprises, Ahmedabad, India

Hydrogen peroxide, tri-sodium citrate dihydrate, citric acid, monobasic sodium phosphate, dibasic sodium phosphate, glycine, hydrochloric acid, acetic acid, non-ionic detergent, all analytical grade, were procured from SD Fine Chemicals or LobaChemie.

Top load washing machine, manufactured by Whirlpool, USA to meet the specification mentioned in AATCC TM135: Dimensional Changes of Fabrics after Home Laundering, and Universal Testing Machine (Testometric, M350-5CT, Rochdale) were used. Both Whirlpool Washing Machine and Testometric Universal Testing Machine were available at OCM Woollen Mills, Amritsar.

2.2 Methods

2.2.1 Treatment of Wool Fabric with Enzymes

The required number of wool samples were treated with enzymes in glycerine bath beaker dyeing machines at liquor ratio 1:20 at specific concentration, *p*H, temperature and time according to the experimental design. Samples were subsequently washed under running water and immersed in boiling water for 15min to deactivate the enzyme. Finally, the samples were washed with cold running water and air-dried. The samples were conditioned for 8h before proceeding with any further testing.

2.2.2 Testing Methods

- Shrinkage of Samples

Total shrinkage of samples was determined using AATCC TM135: Dimensional Changes of Fabrics after Home Laundering. The samples were washed in Whirlpool top loading washing machine with a total wash load of 1.8kg ± 0.1kg. Relaxation shrinkage was observed after 1 wash, and felting shrinkage was observed after total 5 washes. The optimisation of enzymatic treatments was carried out to achieve 3% total shrinkage, as mentioned in Woolmark specification AW-1: 2013 for machine wash care claim. It was observed that the total shrinkage in weft direction was <3% even for untreated wool. For this reason, total weft shrinkage was not considered for optimisation of enzymatic treatment, and thus only total warp shrinkage was considered.

- Tensile Strength

Tensile strength properties of fabric samples were measured in accordance with ASTM D5035 – 06. These values were given as average of 3 values based 1R-T Ravelled Strip Test Method. As total shrinkage in warp direction was considered for optimisation, consequently strength in warp direction only was considered for optimisation.

3 Results and Discussion

3.1 Optimisation of Acid Protease Treatment

Box Behnken design of experiment was used to optimise the treatment parameters of acid protease on Wool. The optimisation was done for concentration of enzyme, *p*H of treatment, temperature of treatment and duration of treatment. Further, the optimisation was based on achieving minimum shrinkage with minimal loss in tensile strength. The details of the experimental setup are given in Table 1 and the results of the experimental runs are given in Table 2.

Table 1 — Experimental setup for optimisation of acid protease treatment and transglutaminase treatment

Parameter	Values	
	AP	TG
Control factors		
Concentration of enzyme, % owf	0.5, 1, 1.5	1, 2, 3
*p*H	4, 5.5, 7	4, 5.5, 7
Temperature, °C	40, 50, 60	40, 50, 60
Time, min	30, 60, 90	30, 60, 90
Response variables	-	-
Total shrinkage in warp direction, % (AP & TG)	-	-
Loss in tensile strength in warp direction, % (AP)	-	-
Strength gain in warp direction, % (TG)		

Table 2 — Experimental results of acid protease treatment on wool and transglutaminase treatment on acid protease treated wool

Exp. No.	Conc., % owf		*p*H	Temp. °C	Time min	Acid protease		Transglutaminase	
	AP	TG				Shrinkage %	Strength loss %	Shrinkage %	Strength gain %
1	0.5	1	4	50	60	5.71	9.5	2.41	5.12
2	1.5	3	4	50	60	2.07	12.21	2.89	7.59
3	0.5	1	7	50	60	6.36	7.35	2.64	4.26
4	1.5	3	7	50	60	3.95	10.38	2.4	6.57
5	1	2	5.5	40	30	6.23	8.05	2.28	3.19
6	1	2	5.5	60	30	5.61	8.55	2.65	5.84
7	1	2	5.5	40	90	3.44	11.3	2.52	4.33
8	1	2	5.5	60	90	2.21	13.33	2.53	7.12
9	0.5	1	5.5	50	30	5.65	6.63	2.53	5.27
10	1.5	3	5.5	50	30	4.13	10.7	2.76	7.03
11	0.5	1	5.5	50	90	3.6	11.17	2.88	5.69
12	1.5	3	5.5	50	90	1.14	14.13	2.76	8.71
13	1	2	4	40	60	5.7	8.32	2.52	3.94
14	1	2	7	40	60	6.39	7.42	2.52	2.06
15	1	2	4	60	60	3.48	10.19	2.52	5.89
16	1	2	7	60	60	5.05	8.05	2.64	3.82
17	0.5	1	5.5	40	60	7.22	7.35	2.64	1.68
18	1.5	3	5.5	40	60	3.54	10.41	2.88	5.11
19	0.5	1	5.5	60	60	4.64	8.19	2.64	4.53
20	1.5	3	5.5	60	60	2.56	12.13	2.52	5.78
21	1	2	4	50	30	5.5	8.57	2.76	6.46
22	1	2	7	50	30	5.46	7.86	3.00	4.38
23	1	2	4	50	90	2.38	13.26	2.52	7.82
24	1	2	7	50	90	3.1	9.72	2.29	5.82
25	1	2	5.5	50	60	2.02	9.849	2.17	8.68
26	1	2	5.5	50	60	2.52	10.061	2.53	8.55
27	1	2	5.5	50	60	2.6	8.954	2.28	8.71
28	1	2	5.5	50	60	2.63	9.778	3.01	7.6
29	1	2	5.5	50	60	1.97	8.482	2.41	7.59

Response surface plots for both total warp shrinkage and strength loss in warp direction are shown in Figs 1 and 2. It can be observed from these plots that there is reduction in shrinkage and higher strength loss with increase in concentration of the enzyme. This can be attributed to increase in damage to both the scales and the internal structure of the wool fibre with increase in concentration of the enzyme, resulting in reduction in shrinkage and increase in strength loss. When *p*H is increased, there is reduction in shrinkage, with minimum shrinkage achieved around *p*H 5. With further increase in *p*H, the shrinkage again increased. In case of strength loss, it is observed that the strength loss kept on increasing with increase in *p*H. This can be attributed to increase in the activity of the enzyme with increase in *p*H till 5. Although with further increase in *p*H the activity of the enzyme decreases, but the strength loss keeps on increasing. This may be because, the enzyme was able to damage the interior structure more as compared to the scales, as scales have higher degree of sulphur crosslinking and harder to digest as compared to the cortex. Similar trends are observed for temperature. Initially, shrinkage is reduced with increase in temperature, with minimum shrinkage observed around 55^{o} C. Further increase in temperature led to some increase in shrinkage. While strength loss increased with increase in temperature till 55^{o} C and beyond this temperature not much increase in strength loss is observed. It can be concluded that the enzyme became more active as temperature increased till 55^{o} C and then remained constant beyond 55^{o} C. For change in time, it is observed that there is decrease in shrinkage and increase in strength loss with increase in time. Further, it is observed that fall in shrinkage is faster as treatment time is increased from 30 min to

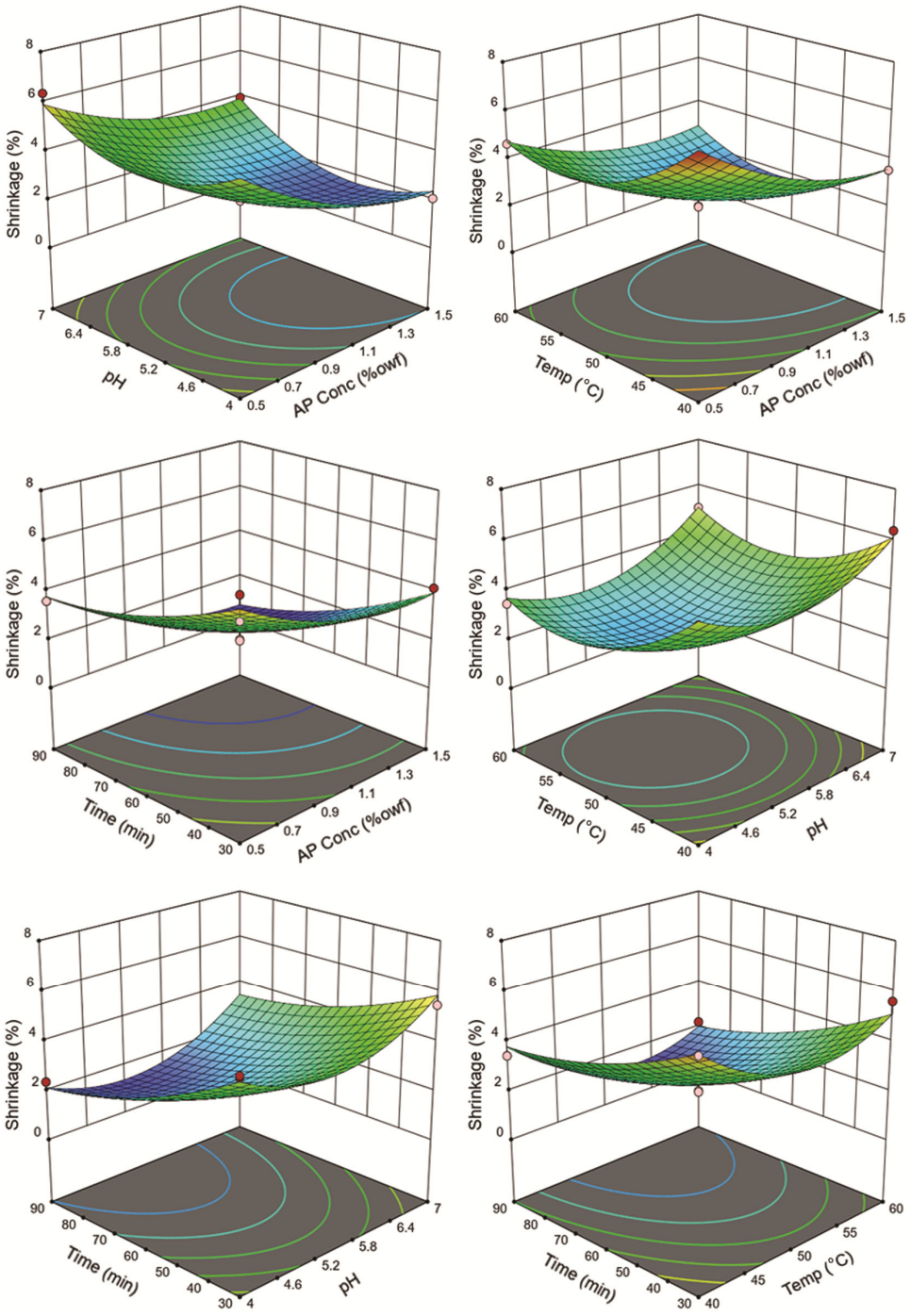

Fig. 1 — Response surface plots for shrinkage after acid protease treatment on wool

60 min, while rate of increase in strength loss is higher when treatment time is increased from 60 min to 90 min. This can be attributed to enzyme acting on the scales more than the internal structure below 60 min and then attacking the internal structure more beyond 60 min. This may be because, the internal structure of wool becomes more accessible to the damage as the time progresses because of enzyme action.

Subsequently, statistical analysis of the data has been carried out and significant terms for the quadratic model are determined. The results so obtained are shown in Table 3. It can be observed from the table that for both the response variables, all

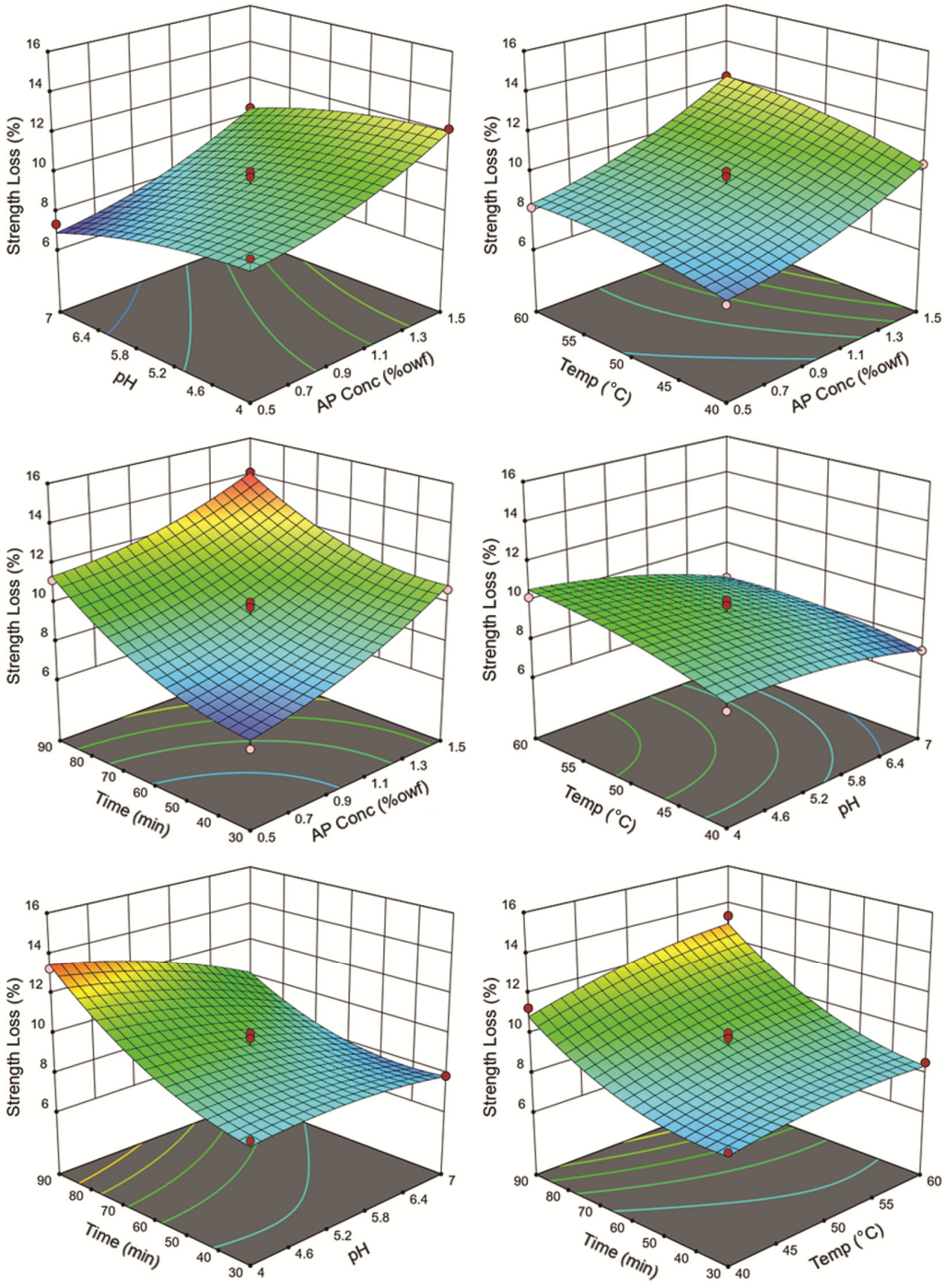

Fig. 2 — Response surface plots for strength loss after acid protease treatment on wool

the control variables are significant model terms. Additionally for shrinkage, squares of all control variables as well as interaction of concentration and temperature are significant model terms, while for strength loss, squares of concentration and time as well as interaction of *p*H and time are significant model term.

Further, statistical optimisation is carried out to provide minimal total shrinkage which should be less than 3% in the warp direction with minimal strength loss in warp direction. Table 4 shows the results of this optimisation. It can be seen that minimum possible total shrinkage achievable from optimisation is 2.54% with 8.76% strength loss. Table 4 also shows

Table 3 — Statistical analysis for optimisation of acid protease and transglutaminase treatments on wool

Response variable	Model	R^2	Adjusted R^2	Predicted R^2	Lack of fit (p-value)
	Acid Protease				
Shrinkage, %	$73.29 - 12.77 \times \text{Concentration} - 6.25 \times p\text{H} - 1.61 \times \text{Temperature} - 0.11 \times \text{Time} + 0.08 \times \text{Concentration} \times \text{Temperature} + 3.07 \times (\text{Concentration})^2 + 0.60 \times (p\text{H})^2 + 0.02 \times (\text{Temperature})^2 + 0.0006 \times (\text{Time})^2$	0.958	0.938	0.902	0.296 (Not significant)
Warp strength loss, %	$3.75 - 1.82 \times \text{Concentration} + 0.32 \times p\text{H} + 0.06 \times \text{Temperature} + 0.01 \times \text{Time} - 0.02 \times \text{pH} \times \text{Time} + 2.56 \times (\text{Concentration})^2 + 0.001 \times (\text{Time})^2$	0.938	0.917	0.889	0.774 (Not significant)
	Transglutaminase				
Strength gain, %	$-98.68 + 8.03 \times \text{Concentration} + 6.69 \times p\text{H} + 3.00 \times \text{Temperature} + 0.09 \times \text{Time} - 0.06 \times \text{Concentration} \times \text{Temperature} - 1.03 \times (\text{Concentration})^2 - 0.66 \times (\text{pH})^2 - 0.03 \times (\text{Temperature})^2 - 0.0005 \times (\text{Time})^2$	0.967	0.951	0.927	0.893 (Not significant)

Table 4 — Predicted and actual values for acid protease and transglutaminase treatments on acid protease treated wool

Parameter	Criteria for statistical optimisation[a]			Concentration % owf	*p*H	Temperature °C	Time min	Shrinkage %	Strength loss/gain %	Desirability
	Shrinkage	Strength loss	Strength gain							
				Acid Protease (AP)						
Predicted	Minimise	Minimise	-	0.97	5.68	50.74	58.15	2.54	8.76 (loss)	0.742
Actual	-	-	-	1	5.7	50	60	2.77	9.95(loss)	
				Transglutaminase (TG)						
Predicted	None	-	Maximise	2.73	5.21	50	65.78	2.59	8.77 (gain)	1
Actual	-	-	-	2.7	5.2	50	65	2.64	7.92 (gain)	

[a]For AP, prediction for optimised treatment parameters was taken by selecting the criteria of minimum shrinkage with minimum strength loss. Similarly for TG, prediction of optimised parameters was taken by selecting 'none' criteria for shrinkage and to maximise the strength gain.

the results for the actual sample when run at the optimised parameters. Figure 3 shows the comparison of total shrinkage and strength of original untreated wool and wool treated with acid protease under optimised conditions. It can be observed that total shrinkage of wool is reduced from 7.66% to 2.77% with 9.95% strength loss.

3.2 Optimisation of Transglutaminase Treatment

Transglutaminase treatment has been carried out on wool treated with acid protease under optimised conditions, as obtained above. Here again, Box Behnken design of experiment is used to optimise the treatment parameters. The optimisation is based on achieving maximum recovery in strength. The details of the experimental setup are indicated in Table 1. The results of the experimental setup are given in Table 2.

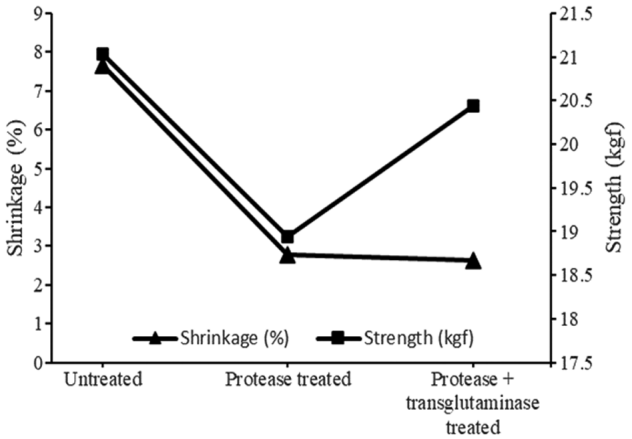

Fig. 3 — Effect of various treatments on shrinkage and strength of wool

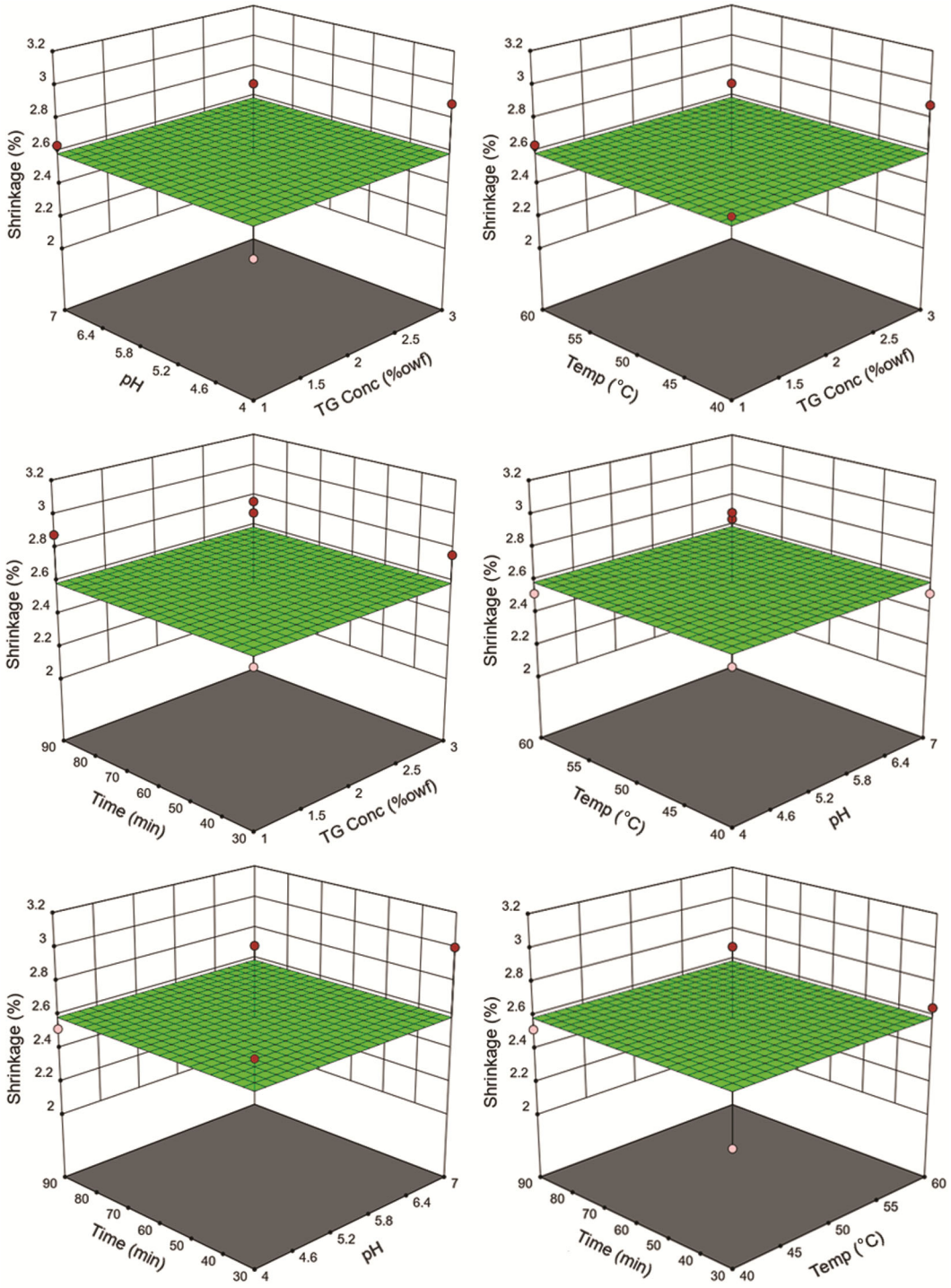

Fig. 4 — Response surface plots for shrinkage after transglutaminase treatment on acid protease treated wool

Response surface plots for both warp shrinkage and strength gain in warp direction are shown in Figs 4 and 5. It is evident from the graphs that transglutaminase treatment has no statistically significant effect on the shrinkage, while all the control factors show influence for strength gain. For this reason, statistical analysis and optimisation are carried out only for strength gain and not for shrinkage. With the increase in concentration, there is improvement in strength gain till 2% (owf) enzyme concentration. But with further increase in concentration, the rate of increase in strength gain

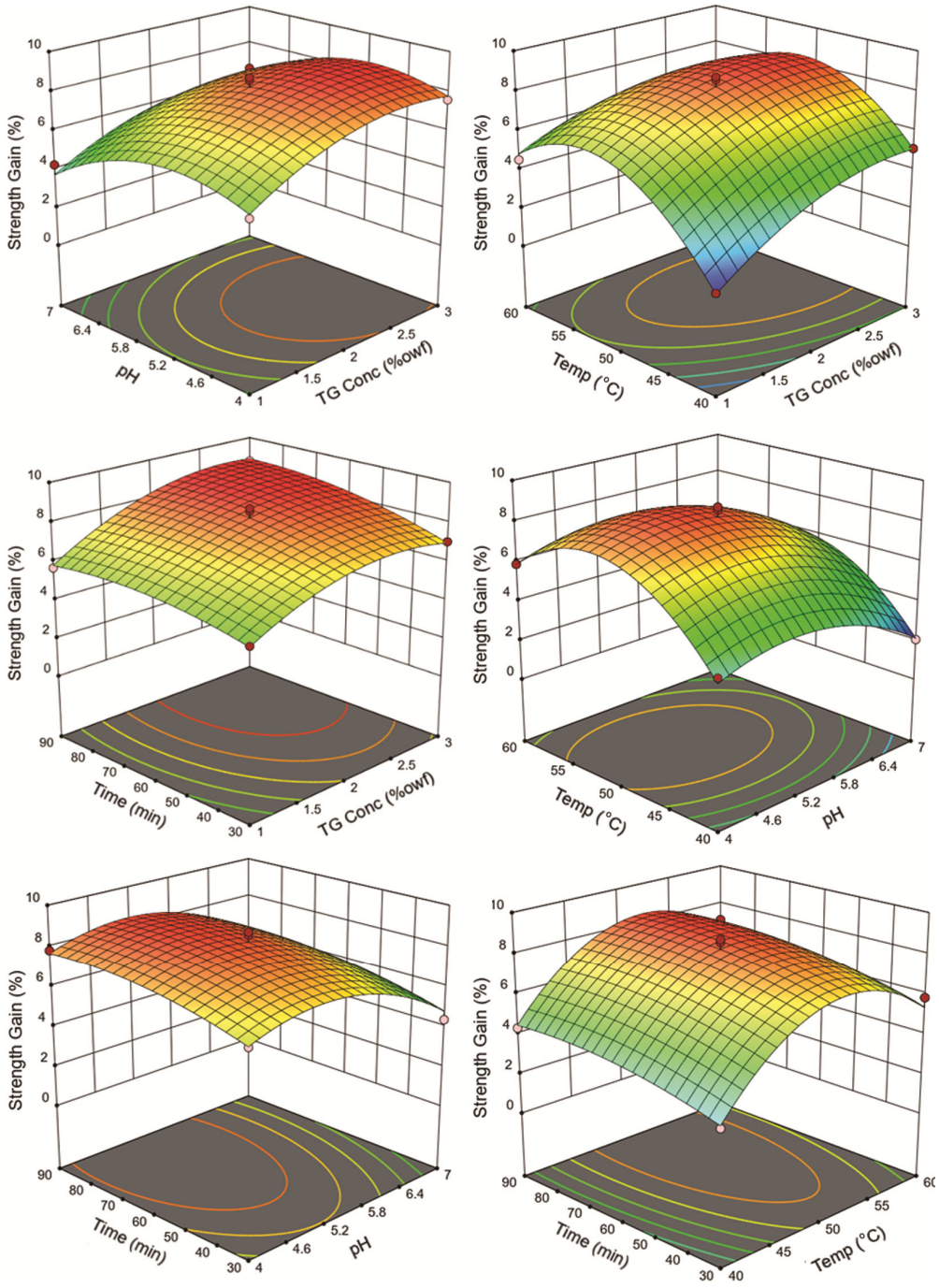

Fig. 5 — Response surface plots for strength gain after transglutaminase treatment on acid protease treated wool

remains quite low. With the increase in *p*H till 5.5, there is slight improvement in strength gain, but the increase in *p*H beyond 5.5 results in fall in strength gain. This can be attributed to inactivation of the enzyme beyond 5.5. Similarly, with increase in temperature from 40ºC to 50ºC, there is considerable improvement in strength gain. Further increase in temperature from 50ºC to 60ºC leads to slight fall in the strength gain. This can be attributed to fall in enzymatic activity beyond 50ºC. With the change in treatment time, there is slight continuous improvement in strength gain with the increase in time of treatment.

As previously done for acid protease treatment, statistical analysis of the data has been carried out to determine the significant terms for the quadratic

model. The results so obtained are shown in Table 3. It can be observed from the table that all the control variables and their squares along with interaction between concentration and temperature are significant for the quadratic model.

Further statistical optimisation is carried out to provide maximum strength gain. The results are shown in Table 4. The comparison of original untreated wool, acid protease treated wool and transglutaminase treated wool is shown in Fig. 3. It can be observed that there is slight reduction in total shrinkage from 2.77% to 2.64%. Also, there is improvement in warp tensile strength giving an increase of 7.92%. When compared to original untreated wool, total warp shrinkage is reduced from 7.66% to 2.64%, while there is a net loss of 2.82% in warp tensile strength.

4 Conclusion

In this study, two different enzymatic treatments conditions are optimised. In the first process, wool is treated with acid protease. In this step, when wool is treated with acid protease using optimised parameters, the total warp shrinkage is reduced from 7.66% to 2.77% with 9.95% loss in warp tensile strength. The optimised treatment conditions are found to be 1% (owf) enzyme concentration, 5.7*p*H, 50^{o}C temperature and 60min treatment time.

In the second process, acid protease treated wool is treated with transglutaminase. When wool is further treated with transglutaminase under optimised parameters, there is marginal reduction in total warp shrinkage to 2.64% with 7.92% recovery in warp tensile strength, giving a net loss of 2.82% in warp tensile strength. The optimised treatment conditions are found to be 2.7% (owf) enzyme concentration, 5.2 *p*H, 50^{o}C temperature and 65min treatment time. Therefore, it is observed that the process is successful in achieving the machine washable care claim as required by Woolmark while minimising the loss in tensile strength.

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